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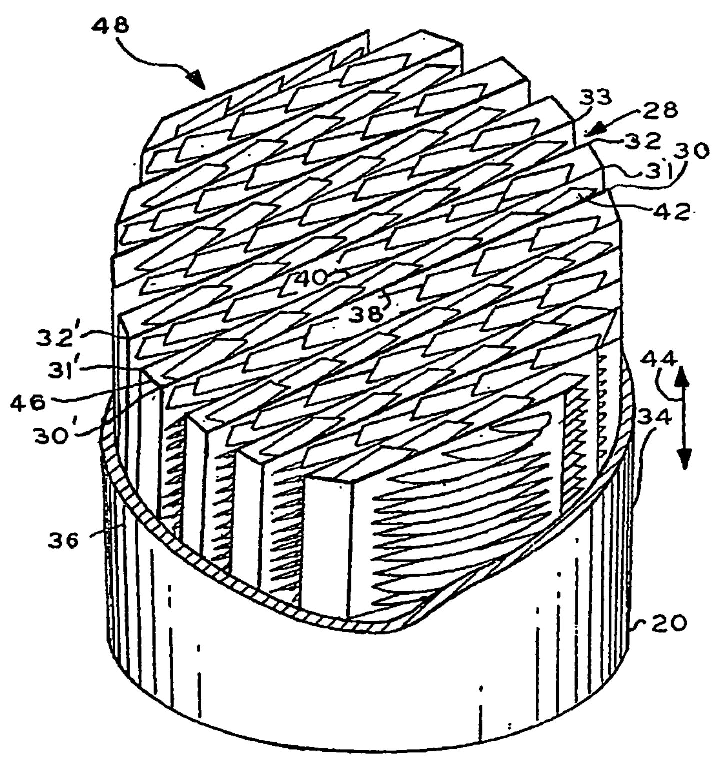
(54) Title: HEAT EXCHANGER/REACTOR APPARATUS



(57) Abstract: A plurality of reaction tubes are in a reaction vessel cavity in which a cooling fluid passes through. A plurality of modular packings are provided in each tube, the packings comprising porous mesh sheet material made of sintered metal fibers having a diameter of about 1-30 microns coated with a catalyst, the material having a void volume of preferably about 80-95%. Each module is formed of a single sheet of material that is folded over at fold lines to form an intermediate interface that thermally conductively engages the tube and a pair of spaced parallel sections, the sections and interfaces forming a zig-zag structure to simplify manufacturing and insertion into the tubes. The sections are planar sheets with turbulence generators in the form of vanes formed from the sheet material and extending into the channels of adjacent sections. End vanes have arcuate edges for directly engaging the tube as do the end edges of the sections and the interfaces to provide enhanced thermal conductive engagement with the tube. The mesh material has a catalyst in the voids thereof. The vanes direct the fluid in the tubes against the sides of the tube bore and also create turbulence for promoting fluid flow through the mesh material.

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HEAT EXCHANGER/REACTOR APPARATUS

This invention relates to catalyst support structures for external heat and mass transport limited chemical processes and heat and mass transport apparatus.

CROSS REFERENCE TO RELATED APPLICATIONS AND PATENTS

Of interest are commonly owned copending U.S. applications Application Serial No. 09/181,186 entitled "Method and Apparatus for Making Catalyst Carrier Device Element" filed October 28, 1998 in the name of Vogt et al., Application Serial No. 09/002539 entitled "Catalyst Carrier Device and Element Therefor" filed January 2, 1998 in the name of Paikert et al. corresponding to PCT/US98/27699 filed December 29, 1998, Application Serial No. 09/156,023 entitled "Coated Products" filed in the name of Schuh et al. on September 17, 1998, Application Serial No. 09/156,023 entitled "Coated Products" filed September 17, 1998 in the name of Schuh et al., Application serial no. 09/ 002,539 filed January 2, 1998 entitled "Structured Packing and Element therefor" in the name of Paikert et al. corresponding to PCT application PCT/US98/27699, application Serial No. 09/181,186 entitled Method and Apparatus for making Structured Packing Element filed October 28, 1998 in the name of Franz Buchi et al. corresponding to PCT/US

99/24907 and Application Serial No. 60/087,474 entitled "Structured Packing and Element Therefor" filed May 29 1998 in the name of Overbeek et al. corresponding to PCT/US99/10784.

Also of interest are U.S. Pats. Nos. 5,304,330, 5,080,962; 5,102,745 and 5,096,663 which disclose making mesh material. All of the foregoing applications and patents are incorporated in their entirety by reference herein.

Catalyst support structures are known for use in heat/mass transfer limited chemical processes in tubular reactors. Catalyst geometries for such processes that are currently performed in small diameter, i.e., less than 8", multi-tubular chemical reactors are chosen to enhance external heat transfer. One type of catalyst support structure is known as Raschig rings. Heat transport is important in highly exothermic reactions and also highly endothermic reactions such as syngas production. The geometry of the catalyst is also important where mass transport is a limiting factor, such as in exhaust gas cleanup.

Catalytic tubular fixed bed reactors are widely used for the production of basic chemicals and intermediates, e.g., the partial oxidation of ethylene to ethylene oxide (single phase, gaseous highly exothermic reaction). The optimum reaction temperature in this case is about 250° C at pressures around 20 bar. Typical industrial reactors are of shell and tube type as shown in Fig. 1. In partial oxidation reactions , the reaction conditions (especially temperature) are critical. If the temperature is too low, the chemical reaction rate is too small and the process becomes more expensive to operate; if it is too high, the sequential reaction of ethylene oxide to carbon oxides is increased at the expense of valuable product.

In an ethylene oxide reactor the catalyst is arranged in tubes and the heat exchange fluid (typically water or oil) circulates externally around the tubes. It is also possible to use a vaporizing fluid as the cooling media. The reactor contains 6,000 to 12,000 tubes of 18 to 38 mm in diameter and up to 10 meters in height. The catalyst is packed in the tubes in the form of ceramic spheres or rings with a diameter of 3 to 10 mm. The reactors are typically run in a recycle mode, since the per pass conversion is low (<50%).

In heterogeneous catalytic reactions heat and mass transfer occurs simultaneously together with the chemical reaction on the catalyst surface. The process can be subdivided into the following steps:

1) mass transport of reactants to the surface by convective and/or diffusion processes.

2) chemisorption of reactants on active centers of the catalyst.

3) chemical reaction

4) desorption of the products

5) mass transport of the products away from the surface.

Some reactions are either strongly exothermic or endothermic; thus, they are very sensitive to the reaction temperature. Therefore, temperature control and heat transfer are major performance issues and the improvement of existing processes depends on their enhancement.

The requirements for the catalyst support can therefore be summarized as:

- efficient heat transfer between catalytic surface and bulk flow and between this bulk flow and the tube wall (heat exchange surfaces).
- efficient mass transfer (for mass transport limited reactions)

- low pressure loss (especially if the conversion/pass is low)
- sufficiently high catalyst concentration and high catalyst utilization – volumetric activity

In addition, the catalyst support structure or packing has to be installed into the tubes and removed from the tubes easily.

Ceramic pellets or spheres are used as catalyst supports in present applications. In a nonadiabatic reaction, the generated heat must be transported perpendicular to the main flow direction in the tubes through the fixed catalyst bed to the tube walls. In a dumped packing of pellets, the radial heat transport is established as a result of the random arrangement and the resulting tortuous flow around the pellets. Hollow and full cylinders are in this case more effective than spheres due to their greater non-uniformity. The heat transport at the boundary between the fixed bed and the tube wall is also decisive. The flow within the tube is not uniform with higher velocity along the tube wall than in the center due to differences in free volume (flow resistance). Thus, the flow will bypass the center of the packing and heat transfer from the tube center to the wall is reduced. This velocity profile is established within the first 10-20 equivalent packing diameters, creating a barrier for effective convective radial heat transfer.

The present inventors recognize a need for an improved tubular reactor to create a more uniform radial temperature profile within the heat exchanger tubes to provide more uniform reaction conditions leading to higher reaction selectivity and which will overcome the problems noted above with the prior art heat exchanger catalyst support or packing structures. More particularly, the present inventors recognize that catalyst coated micro fiber metal sheets will provide both an

improvement in volumetric catalyst activity, particularly due to a decrease in mass transfer resistances, and also the desired more uniform radial temperature profile within the heat exchanger tubes. The problem with such micro fiber sheets is the need to place such material in small diameter tubes, i, e., provide ease of manufacturability, while at the same time provide enhanced heat or mass transfer. Therefore, such sheets need to be appropriately configured to be easily placed in small diameter tubes.

A heat/mass transfer apparatus according to the present invention comprises a thermally conductive tube having a longitudinal axis for receiving a first fluid internally thereof thermally conductively contacting the tube internal surface and having an external surface for transfer of heat energy from the internal surface to a second cooling fluid thermally conductively contacting the tube external surface.

A packing is included and comprises at least one catalyst coated porous thermally conductive sheet material in the tube, the sheet material having pores exhibiting a plurality of interstices in fluid communication with each other and externally the material, the sheet material for providing a reaction with the first fluid and for the transfer of reaction heat energy in the first fluid to the tube, the sheet material lying in a plane extending along the tube axis and including vanes extending therefrom arranged for directing the first fluid laterally across the tube axis against the tube for optimizing energy transfer from the first fluid to the tube and for thermally conductively transferring energy from the sheet material to the tube.

In one aspect, the sheet material comprises a plurality of sections, adjacent sections forming a fluid passage chamber therebetween and extending in the axial

direction of the tube axis. The vanes form turbulence generators and extend into the corresponding chamber from the sections and are arranged for directing fluid flow laterally along the corresponding section relative to the tube axis toward the tube.

The catalyst is secured to the material for reacting with the fluid as the received first fluid impinges on the packing and passes through the interstices.

The porous material preferably comprises sintered metal fibers.

In a further aspect, the porous material comprises planar sheet material.

In a further aspect, the packing is modular, a plurality of said modular packings being located in the tube in series along the tube axis.

In a still further aspect, the packing is a module comprising a plurality of sheet material sections, adjacent sections defining a fluid channel therebetween.

In a further aspect, the packing is a single sheet of material with a plurality of sections, each section being connected by fold lines to an adjacent section.

In a further aspect, the adjacent sections are interconnected by an intermediate interface.

Preferably each section has opposing edges in thermal conductive contact with the tube.

In a further aspect, the sections are spaced to form a channel therebetween, adjacent sections being connected by an interface in thermal conductive contact with the tube.

The turbulence generators preferably comprise an array of vanes extending from each section, the array being aligned along the tube axis.

In a further aspect, the vanes have a width dimension and a length dimension that are inclined relative to the tube axis, the length direction being inclined laterally the tube axis to direct the first fluid against the tube.

IN THE DRAWING:

FIGURE 1 is a front elevation schematic sectional view of a heat/mass transfer apparatus according to an embodiment of the present invention;

FIGURE 2 is an isometric view of a packing module used in the tubes of the apparatus of FIGURE 1;

FIGURE 3 is a further isometric view of the packing module of Fig. 2;

FIGURE 4 is a top plan sectional view of the a tube of the embodiment of Fig. 1 showing a packing module in a tube;

FIGURE 5 is a plan view of a set of blanks used to make the modules of Figs. 2 and 3;

FIGURE 6 is a more detailed view of a portion of one of the blanks of Fig. 5 illustrating the vane formation;

FIGURE 7 is an isometric schematic view of a tube with a further embodiment of a catalyst support structure therein; and

FIGURES 7a, 7b and 7c are sectional plan views through the embodiment of Fig. 7 taken at different axial locations of the tube.

In Fig. 1, a reaction vessel 2 comprises a metal, e.g., stainless steel, shell 4 having a cavity 6. The shell 4 is a circular cylinder with semi-spherical end plates 8 and 10 at the respective top and bottom of the cavity 6. A gas inlet pipe 10 is at the bottom end plate for supplying a gas to be processed into inlet manifold 16. A gas outlet pipe 14 receives processed gas from exhaust manifold 18.

An array of linear tubes 20, which may be metal or other good heat conductive material, and which may be of any desired internal diameter as discussed in the introductory portion, are connected in parallel in fluid communication with the inlet and exhaust manifolds 16 and 18. Thus fluid flowing into inlet manifold 16 from the pipe 12 flows through the tubes 20 into the exhaust manifold 18 into the outlet pipe 14. The structure thus far described is available in the prior art.

The cavity 6 has a cooling fluid inlet 22 and a cooling fluid outlet 24. Cooling fluid such as water or other liquid or gas flows into the cavity in convective contact with all of the tubes 20. Vaporizing liquid can also be used for cooling, just as condensing fluid can be used if heating is necessary. The cooling fluid transfers heat from the tubes 20 generated by the reaction in tubes 20. The cooling fluid, e.g., water, cools the reaction fluid, e.g., gas., flowing in the tubes 20 from the gas inlet pipe 12 to the outlet pipe 14. As mentioned in the introductory portion, prior art reactors/heat exchangers employ ceramic rings or other structures in the tubes as catalyst support structures. As described these structures are not as efficient as desired for controlling the temperature of the fluid flowing in the tubes 20 in the desired temperature range and, therefore, the reactions in the tubes are not as efficient as desired. The catalysts may not provide as complete a reaction as desired or excessive amounts of other unwanted products may be made.

The term mass transfer in this art refers to the motion of the fluid and its contact with the catalyst. As mentioned in the introductory portion, the mass transfer refers to the transport of reactants to the surface of the catalyst by convection or diffusion, chemisorption of the reactants to the centers of the catalyst in order to insure complete reaction with the catalyst, the chemical reaction itself,

the desorption of the products and the mass transport of the products away from the surface. The prior art support structures and catalysts thereon do not provide efficient processing in respect of these various parameters involved with the chemical reaction process as described in the introductory portion. The present invention provides modules of catalyst support structure packing inside the tubes which are significantly more efficient in transferring heat / unit of pressure loss.

In Figs. 2, 3 and 4, a catalyst support structure or heat transfer modular packing 28, according to a given implementation, is placed axially in each tube for the length of each tube 20. The packings 28 each comprise a single one piece sheet of porous mesh or screen material made of metal or other fibers. The fiber material may also be ceramic, glass, carbon or any combination thereof. The modular packings 28 are place in preferably abutting (or closely spaced relation) in the tube 20 bore.

Representative modular packing 28 comprises a single sheet of the porous mesh material. The mesh material, Fig. 4, is folded at fold lines 30, 31, 33 and so on at one side 34 of the packing 28, and at fold lines 30', 31' and 33' and so on at the opposite side of the 36 tube 20. Fold lines 30, 30' define a planar section 38 therebetween of the flat planar sheet mesh material. Fold lines 31, 31' form an adjacent planar section 40 of mesh material. Sections 38 and 40 form a fluid flow channel 42 therebetween for fluid flowing nominally in direction 44 therein from the inlet manifold 16 to the outlet manifold 18, Fig. 1. The actual direction of fluid flow in the tubes is complex due to turbulence as will be described and also flows inclined transverse to the tube longitudinal axis defined by direction 44.

The region between sections 38 and 40, by way of example, between fold lines 30' and 31' forms a generally rectangular intermediate tube interface 46 which abuts the tube 20 inner surface. The sections 38 and 48 which are representative of the orientation of the other sections in the packing 28 are parallel and parallel to the other sections in the packing. As a result there is an array 48 of parallel sections, each section terminating at a foldline forming an interface with the fold line of the adjacent section. The intermediate interfaces such as interface 46 all abut an inner surface of the tube 20 in preferable thermal conductive relation. The sections such as sections 38 and 40 and so on are all interconnected as a one piece structure separated by fold lines and an intermediate interface, such as interface 46.

The array of sections such as sections 38 and 40 form a corresponding array of fluid channels such as channel 42 which are all parallel of generally the same transverse width in directions 50, Fig. 4. Located in each channel are turbulence generator vanes 52, 54 and 56, for example in channel 58. The vanes are all inclined at about 45° with respect to the fluid flow direction 44 through the tube 20, but may be inclined at other angles. The vanes redirect fluid impinging on the vanes transversely against the tube 20 inner side wall surface to optimize heat transfer to the tube. The vanes 52, 54 and 56 are just a few of the vanes attached to section. Other like vanes are in spaced alignment with the vanes 53, 54 and 56 in the axial fluid flow direction 44 of the tube 20 in a vertical array. Either an interface or vane (at the edge of the packing such as vanes 57 and 59, Fig. 4) is in thermal conductive contact with the inner surface of tube 20. The modular packing 28 is thus a zig-zag structure folded in accordion fashion with somewhat rectangular channels formed by planar sections and intermediate interfaces. The intermediate interfaces are at angles

to the plane of some of the sections so as to mate with corresponding curvature of the tube 20 inner surface as shown in Fig. 4.

The configuration and layout of the vanes 52, 54, 56 and so on is best illustrated in connection with figures 5 and 6. In Figs. 5 and 6, the orientation of the vanes are different, but the dimensioning of the vanes is the same for a given tube internal diameter as the relative orientation of the vanes is not critical for a given tube, the orientation of all of the modules preferably being the same in each corresponding tube. However, the orientation of the vanes, which may be about 45° to the longitudinal axis of the tube may also be different for a given set of modules in a tube according to a given implementation.

In Fig. 5, three identical rectangular blank sheets 62, 62' are formed of wire mesh from a blank 63, the mesh material to be described below. Representative sheet 62' is an elongated rectangular sheet of fiber mesh material having two parallel identical longitudinal edges 64 and parallel identical end edges 66. Solid lines in the blank 63 sheet represent through cuts. The blank sheet 62' has a plurality of aligned sections 68, 70 and 72 and so on in a linear array. The sections have different lengths L that corresponds to the transverse dimension across the tube 20 internal diameter for that section (see Fig. 4). The interfaces are between each such section such as interfaces 74, 76 and 78. The interfaces alternate on opposite sides of the tube 20 as shown in Fig. 4. The vanes are formed by cuts 88, Fig. 6, in section 104 at 45° to the length dimension of the blank and sections from left to right in the figure.

As best seen in Fig. 6, in blank 104 the vanes such as vanes 80, 82 and 84 in representative section 86 are identical and formed by through cuts 88. Vanes 90 and

92 are shorter than vanes 80, 82 and 84 as they are located in the corner of the section. The mirror image vanes 94 and 96 in the diagonal opposite corner of section 86 are the same as vanes 90 and 92, but in the alternative may differ from each other according to a given implementation.

Cut 88 has a straight portion 88' and an angled cut 88" at one end of the cut and a U-shaped cut 98 in conjunction with cut 88'. Representative vane 84 has a fold line 100 shown by the dashed line. The fold lines for the vanes in the other sections are not shown by dashed lines, but are intended to be included. The fold lines for all of the central sections in blank 104 excluding the two opposite end sections such as section 102 are parallel to fold line 100.

The sections are each separated by two fold lines such as fold lines 106 and 108 between sections 86 and 102. Sections 106 and 108 form intermediate interface 110 therebetween. A further intermediate interface 112 is between fold lines 114 and 116 of respective sections 118 and 86 and so on.

The vanes of end section 102 are different then the vanes intermediate the end sections. The vanes 120, 122, 124,126 and so on of the end section 102 are thinner in transverse width, and have curved external edges 128. These vanes directly abut the inner surface of the tube and therefore have curvatures that match the curvature of the curved inner surface of the tube 20. These end section vanes correspond in location to vanes 54, 57, for example, in Fig. 4, modular packing 28. It should be understood that the drawings are not to scale and are generally schematic in nature to explain the principles rather than provide exact dimensional relation of the different elements of the packing and tube 20.

Because the vanes of the different modular packings 28, Figs. 2-4, are inclined generally at 45° to the longitudinal axis of the tubes 20, these vanes all direct fluid against the inner surface of the tube walls to maximize heat transfer from the interior of the sections to the tubes . The vanes also create local pressure differentials, i.e., turbulence, needed to maximize fluid flow through the mesh of the substrate material forming the modular packing 28 as will be described in more detail below. The mesh material because of the small pore size normally does not exhibit fluid flow therethrough when the pressure differential thereacross on opposite surfaces is about the same or a small value.

The mesh material forming the packing 28 used throughout the apparatus 2 in all of the tubes is of the same construction. The mesh or screen which is a micromesh is used as the porous packing. The mesh material is a three-dimensional network of fibers or wires, with such fibers or wires generally having a diameter of at least 1 micron and a diameter which generally does not exceed 25 microns, although smaller or larger diameters may be employed. The network may be of the type described in U.S. Patent Nos. 5,304,330; 5,080,962; 5,102,745; or 5,096,663. The three-dimensional network of materials may be one which is comprised of fibers, and may be a felt or the like, a fiber filter or paper and the like suitable for high temperature processes employed in reaction systems of the type described, or may be a porous composite. The compacted wires or fibers define a three-dimensional network of material which has a thickness thereto. In general, the thickness of the three-dimensional network of material is at least 5 microns, and generally does not exceed 10 mm. In general, the thickness of the network is at least 50 microns and does not exceed 2 mm.

The three-dimensional network may be coated or uncoated with a catalyst and such three-dimensional network may have particles entrapped or contained therein. The network may have different pore sizes over the thickness thereof and may be laminated and/or comprised of the same materials and/or may have multi-layers.

It is to be understood that the mesh may be comprised of one type of fiber or may be comprised of two or more different fibers or the mesh fibers may have a single diameter or may have different diameters. The mesh is preferably formed of a metal, however, other materials may be employed such as a ceramic. As representative examples of such metals, Nickel, various stainless Steels; e.g., 304, 310, and 316, Hastelloy, Fe-Cr alloys, and so on may be used. The mesh can retain particles or fibers in the interstices thereof and the particles or fibers contain a catalytic function.

The modular packing preferably includes a catalyst for reaction processes in this embodiment. The catalyst, if used, may be coated on the fibers forming the packing and/or supported or unsupported catalyst may be entrained in the mesh openings.

Although it is preferred to fabricate the packing 28 from porous materials such as a micromesh structure, Applicants note that in order to efficiently use such porous materials as packings, it is necessary to provide turbulence generators which are spaced over the packing structure in order to provide for efficient flow of liquid through pores in the packing. That is, as mentioned above, the flow of fluid, gas or liquid does not normally occur through the material unless a pressure differential is created thereacross of sufficient magnitude so as to promote fluid flow therethrough.

In a preferred embodiment, in addition to the turbulence generators, the packing can be provided with additional openings. These openings are provided by the relatively large regions produced by the vanes as they are folded out of the plane of the sections of the modular packing 28.

The invention has been described with respect to a representative embodiment of modular packing structure formed from a mesh material; however, such structures are by way of illustration in that the present invention is applicable to other structures and designs. It is recognized herein that highly porous mesh material, when used as packing, even though such material has a high-void volume; for example, greater than 70% and in many cases greater than 90%, e.g., 95%, fluid does not effectively flow through the pores of the packing and that fluid flow through such pores can be improved by providing turbulence generators. Thus, the vanes form both turbulence generators and fluid flow directivity to the tube walls.

The vanes direct fluid flow laterally across the tube internal diameter transverse the longitudinal tube axis. The widths of the vanes are inclined in the axial direction of the tube axis and the length dimension of the vanes is inclined in the transverse lateral direction to direct and channel fluid flow to the tube inner side wall surfaces. The vanes create turbulence in the fluid attempting to flow vertically in the chambers along the tube axis. The fluid also flows transversely the tube axis to and between the chambers through the openings in the section walls as the fluid is directed transversely toward the tube side walls.

The size and spacing of the openings in the mesh material of the sections, preferably in combination with the vane turbulence generators, are selected to obtain a desired bulk mixing and pressure drop through the mesh of the structured packing.

The mesh-like structure may be comprised of a single layer, or may include multiple layers where there may be one layer of wires; e.g., a knitted wire structure or a woven wire structure, and preferably comprises a plurality of wires or fibers to form a three dimensional network of materials. In one embodiment, the support structure comprises a plurality of layers or sheets of fibers that are oriented randomly in the layers. One or more metals may be used in producing a metal mesh. Alternatively the mesh fibers may be formed from or include materials other than metals alone or in combination with metals; e.g. carbon or metal oxides or a ceramic.

In general, the thickness or diameter of the fibers which form the plurality of layers of fibers is preferably less than about 500 microns, more preferably less than about 150 microns and more preferably less than about 30 microns. In a preferred embodiment, the thickness or diameter of the fibers is from about 1 to about 30 microns and preferably 8-25 microns.

The three dimensional mesh-like structure may be produced as described in U.S. Pat. Nos. 5,304,330, 5,080,962; 5,102,745 or 5,096,663 incorporated by reference herein. It is to be understood, however, that such mesh-like structure may be formed by procedures other than as described in the aforementioned patents.

The term "void volume" as used herein is determined by dividing the volume of the structure which is open by the total volume of the structure (openings and mesh material) and multiplying by 100. The particular description of the mesh-like sheet material is also described in the aforementioned Application Serial No. 60/116,649 filed January 21, 1999 and incorporated in its entirety by reference herein.

The sheet of mesh material is coated with a catalyst. The supported catalyst which is supported on the mesh-like structure may be present on the mesh-like support as a coating on the wires or fibers that form the mesh-like structure and/or may be present and retained in the interstices of the mesh-like structure.

In one embodiment, wherein the catalyst supported on a particulate support the catalyst is present as a coating on the mesh-like structure, the mesh-like structure may be initially coated with a particulate support, followed by addition of the catalyst to the particulate support present as a coating on the mesh-like structure. The particle support for the catalyst may include alumina, silica, zirconia, ceria, titania, barium oxide and mixtures thereof. Examples of specific compositions are disclosed in the aforementioned Pat. Nos. 4,9065,243; 4,939,113, and PCT Application No. WO 95/35152. Alternatively, the catalyst supported on a particulate support may be coated onto the mesh. The particulate support with or without catalyst may be coated on the mesh structure by a variety of techniques, e.g., dipping or spraying. After coating the particulate support without catalyst onto the mesh, the particulate support is impregnated with a solution containing the catalyst precursors and is treated thermally to obtain the catalyst.

In one embodiment, wherein the mesh-like structure is comprised of a plurality of layers of metal fibers, the particulate support with or without catalyst may be coated onto the mesh-like catalyst support by an electrophoretic coating procedure, as described in U.S. Application Serial Number 09/156,023, filed on September 17, 1998 incorporated by reference herein. In such a procedure, a wire mesh-like structure is employed as one of the electrodes, and the particulate support, such as an alumina support of the requisite particle size, with or without catalyst, (which may

include alumina in the form of a sol to promote the adherence of larger particles to the wire mesh) is suspended in a coating bath. A potential is applied across the electrodes, one of which is the mesh-like structure formed from a plurality of layers of fibers, and the mesh-like structure is electrophoretically coated with the alumina support with or without catalyst. If the alumina support does not include a catalyst, the catalyst then can be added to the catalyst structure by impregnating with or dipping the structure (which contains the alumina coating) into an appropriate solution that contains the catalyst and possibly one or more promoters.

As hereinabove indicated, the supported catalyst may be supported on the mesh material by entrapping or retaining the particulate support in the interstices of the mesh. For example, in producing a mesh-like structure comprised of a plurality of layers of randomly oriented fibers, the particulate support may be included in the mix that is used for producing the mesh-like structure whereby the mesh-like structure is produced with the particulate support retained in the interstices of the mesh. For example, such mesh-like structure may be produced as described in the aforementioned patents, and with an appropriate support being added to the mesh that contains the fibers and a binder, such as cellulose. The catalyst or support, entrapped within the interstices of the metal fibers and the binder is subsequently, heated in a reducing atmosphere such as hydrogen such that the fibers attach to each other by sintering action at the fiber contact points within the porous material. In the alternative, the metal fibers and catalyst or support may be heated by flowing an electric current through the material in a flowing gas atmosphere so as to cause the fibers to attach to each other at fiber contact points within the porous material. In the case the produced mesh structure includes the particulate support retained in the mesh structure, the particulate

support retained in the mesh structure is impregnated with the catalyst precursors and treated thermally to obtain the catalyst.

In one example, the fibers are about 8-12 microns in diameter and are employed in sheets of about 0.5 mm thick with 95% voids. The material has the consistency of paper or felt, is delicate and can be readily permanently deformed and/or damaged. The sheets are 1m x 1m and corrugated as described above. These sheets are cut to size for corrugating the material. The material is then cleaned in a solvent, e.g., acetone, to remove contamination and dried in heated air. Optionally, the material may be heat treated at 400-700°C in air to preoxidize the surface of the metal fibers. The catalyst coating is applied in a slurry process, a catalyst carrier or wherein the catalyst is suspended in water (colloidal suspension, slip) and the metal fiber sheet is immersed into the slurry. Because the slurry must penetrate the entire mesh structure (coat the fibers in the void volume), the wetting behavior must be appropriate. Heat treatment provides good wetting properties because a slight oxidation layer on the fibers provides the desired wetting action. However, depending on the material, wetting can be acceptable without heat treatment in accordance with the manufacturing process of the fiber sheets and the fiber wetting characteristics. After heat treatment, contamination needs to be avoided with careful handling of the sheet material.

The coating is applied in the slurry containing the catalytic powder. The slurry contains the catalytic powder of a composition as described in the catalyst related patents and International Application noted in the introductory portion. Preferably, this slurry is deposited as a thin coating, e.g. 1-10 microns, onto the micro fibers. This deposition can be by an electrophoretic process described more

fully in copending Application Serial No. 09/156,023 noted in the introductory portion and incorporated by reference herein. After the electrophoretic coating is applied, the sheets are dried in hot air and heat treated to provide bonding of the catalyst coating on the fibers. The sheet material is heated to about 200-900°C during this heat treating in accordance with the nature of the coating. For example, when a catalyst is directly deposited on to the fibers, a heat treatment of 500°C may be employed. This provides the desired attachment of the coating onto the fiber surfaces and may not affect the properties of the catalyst.

After final heat treatment, the single sheets are formed.

The preferred micro fiber mesh material which can be provided by the sintered fiber sheet material of the catalyst carrier device elements provides relatively high catalyst surface area with optimum access to the catalyst by fluids. Where relatively rapid chemical reactions are employed, utilization of the internal surface area of the porous material is dependent upon the rate of transport of the gases to these surfaces. The mass transport is faster in the case of driven forced flow (convection) than by mere concentration of gradients (diffusion). The intended optimum device, therefore, provides optimum cross flow of the fluids at low pressure differentials there across.

To optimize reaction efficiency, the pressure drop is maintained relatively low as discussed above. This is provided by relatively high void space per unit volume, low friction (good aerodynamic characteristics) and prevention of undesirable stagnant gas pockets.

In Fig. 7, an alternative embodiment of a modular packing is shown and includes a tube 130 with the packing 132 therein. The packing 130 includes a

support rod 134 and mesh elements 136 and 138. Elements 136 are all identical and comprise a flat sheet of mesh material as described above. In Fig. 7a, for example, element 136' comprises a segment of a circular disc having a linear chord edge 138 forming a gap 140 with the tube 130.

In Fig. 7b, element 136" edge 138' forms a gap 140' with the tube inner surface and is rotated 90° from the orientation of the element 136'. Similarly in Fig. 7c, element 136'" has an edge 138" forming a gap 140" with the tube inner surface. All of the elements are supported by circular cylindrical rod 134 passing through the element centrally thereof. The element 136'" is rotated 90° from the orientation of element 136" or 180° from that of element 136'. A fourth element not shown is rotated 180° from the orientation of that of the element of Fig. 7b. This staggered relationship is repeated throughout the length of the rod 134. The elements are bonded to the rod in the different 90° angular orientations about the rod as seen in the figures.

The ends of the rod 134 are bent to form a J-hook 142. The modular packings 28 and 132 can easily be inserted into the tube 130 bore. The modular packing 28 of Figs. 2-4 is preferred as this is the simplest to manufacture. Other modules are inserted serially to fill the tube. The staggered relationship of the elements of Fig. 7 provides the desired pressure drop and turbulence and also flows the fluid in the tube against the side walls to optimize the heat transfer to the tube.

It can be shown that the modular packings described herein with the described mesh structure provides optimum heat transfer to the tubes to maintain the desired temperature of the fluid flowing in the tubes. Also, this mesh material provides optimum reaction of the fluids with the catalyst on the mesh material.

Still other vane or element structures may be provided a modular packing in accordance with the principles described herein. It will occur to one of ordinary skill that various modifications may be made to the disclosed embodiments which are given by way of illustration and not limitation. It is intended that the appended claims define the invention.

What is claimed is:

1. A heat/mass transfer apparatus comprising:

a thermally conductive tube having a longitudinal axis for receiving a first fluid internally thereof thermally conductively contacting the tube internal surface and having an external surface for transfer of heat energy from the internal surface to a second cooling fluid thermally conductively contacting the tube external surface; and

a packing comprising at least one porous thermally conductive sheet material in the conduit, the sheet material having pores exhibiting a plurality of interstices in fluid communication with each other and externally the material, the sheet material for providing a reaction with the first fluid and for the transfer of reaction heat energy in the first fluid to the tube, the sheet material lying in a plane extending along said tube axis and including vanes extending therefrom arranged for directing the first fluid laterally across the tube axis against the tube for optimizing energy transfer from the first fluid to the tube and for thermally conductively transferring energy from the sheet material to the tube.

2. The apparatus of claim 1 wherein said sheet material comprises a plurality of sections, adjacent sections forming a fluid passage chamber therebetween and extending in the axial direction of the tube axis, the vanes forming turbulence generators and extending into the chamber from the sections and arranged for directing fluid to flow laterally relative to the tube axis toward the tube.

3. T pparatus of claim 1 including a catalyst secured to the material for reacting with said fluid as the received first fluid impinges on said packing and passes through the interstices.

4. The apparatus of claim 1 wherein the porous material comprises sintered metal fibers.

5. The apparatus of claim 1 wherein the porous material comprises any one or more of the group consisting essentially of metal, ceramic, glass or carbon fibers.

6. The apparatus of claim 1 wherein the porous material comprises planar sheet material.

7. The apparatus of claim 1 including a catalyst secured to the material within said interstices.

8. The apparatus of claim 1 wherein the vanes are parallel to each other and are inclined to the longitudinal axis of the tube.

9. The apparatus of claim 2 wherein the vanes comprise a planar sheet material element extending at an angle to the plane of the corresponding section for about the width of the corresponding section in a direction transverse the tube axis and inclined to the axis in a lateral direction transverse the longitudinal axis of the tube.

10. The apparatus of claim 1 wherein the packing is modular, a plurality of said modular packings being located in the tube in series along the tube axis.

11. The apparatus of claim 1 wherein the packing is a module comprising a plurality of sheet material sections, adjacent sections defining a fluid channel therebetween.

12. The apparatus of claim 1 wherein the packing is a single sheet of material with a plurality of sections, each section being connected by fold lines to an adjacent section.

13. The apparatus of claim 10 wherein the adjacent sections are interconnected by an intermediate interface.

14. The apparatus of claim 12 wherein each section has opposing edges in substantial thermal conductive contact with the tube.

15. The apparatus of claim 12 wherein the sections are spaced to form a channel therebetween, adjacent sections being connected by an intermediate interface in thermal conductive contact with the tube.

16. The apparatus of claim 12 wherein the vanes comprise an array of vanes extending from each section, the array being aligned along the tube axis, a plurality of the vanes extending to a region adjacent to the tube opposing inner wall surfaces.

17. The apparatus of claim 16 wherein the vanes are inclined relative to the tube longitudinal axis in a lateral direction transverse the tube axis to direct the first fluid against the tube.

18. The apparatus of claim 16 wherein the vanes extend transversely across the internal diameter of the tube and are spaced from the next adjacent section.

19. The apparatus of claim 12 wherein the vanes extend transversely across the internal diameter of the tube, the vanes of each section being aligned in the same plane along the tube axis.

20. The apparatus of claim 13 wherein the vanes are rectangular in the channels having an interface and have arcuate edges in channels formed by a section on one channel side and a tube inner surface on an opposing channel side.

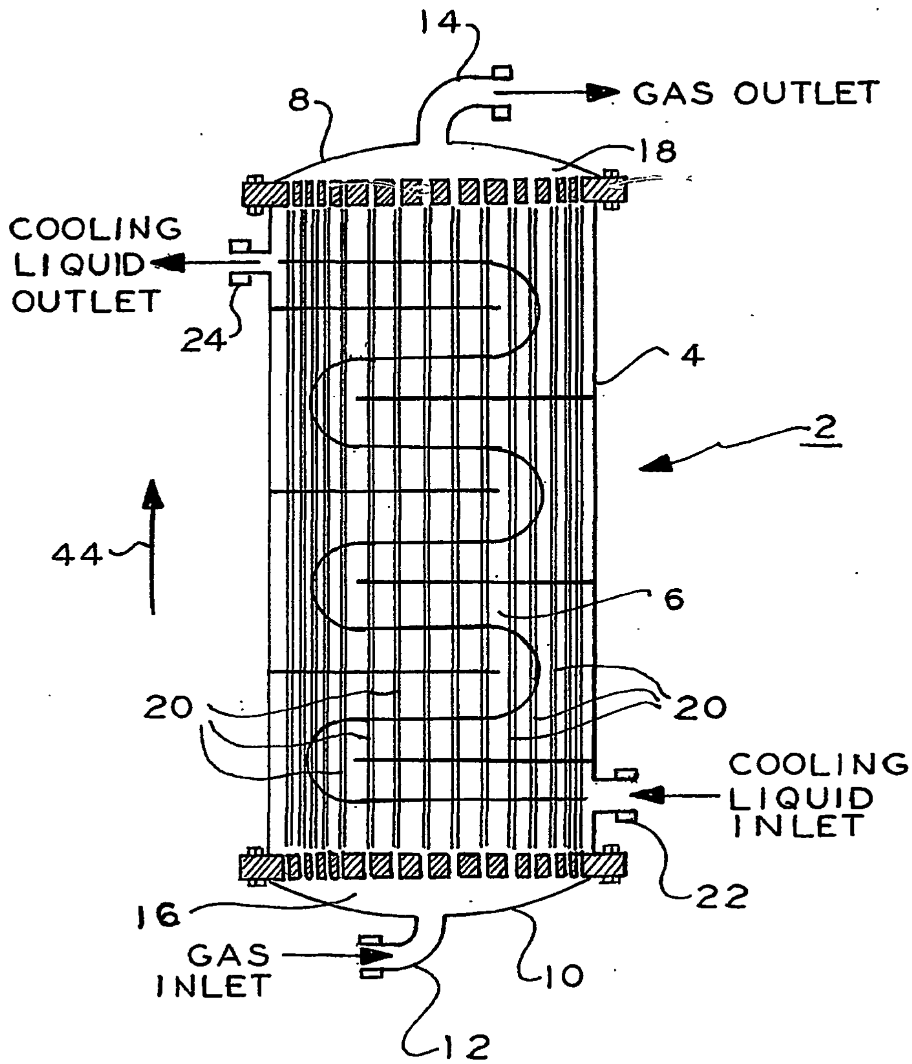
21. The apparatus of claim 12 wherein the vanes are formed from the sheet material forming through openings in the sheet material corresponding to the vane dimensions.

22. The apparatus of claim 1 wherein the packing is formed into a module, a plurality of modules being arranged in an axial array in the tube.

23. The apparatus of claim 1 wherein the tube internal diameter is no greater than about 6 inches (about 15 cm).

24. The apparatus of claim 1 wherein the tube internal diameter is less than one inch (about 2.5 cm)

FIG. 1



14
GAS OUTLET
8
18
COOLING LIQUID OUTLET
24
4
2
44
6
20
20
COOLING LIQUID INLET
22
16
GAS INLET
10
12

FIG. 2

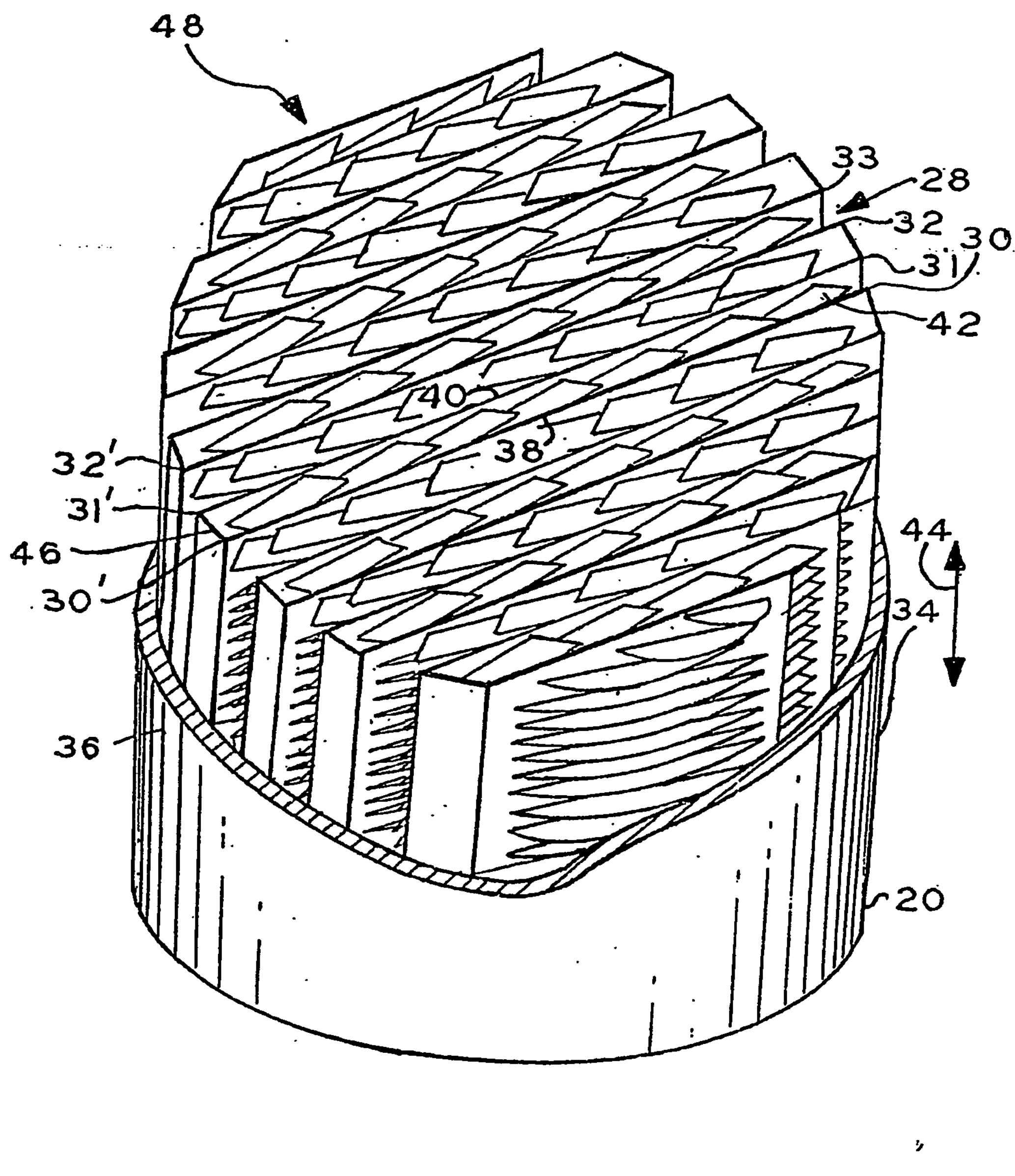


FIG. 3

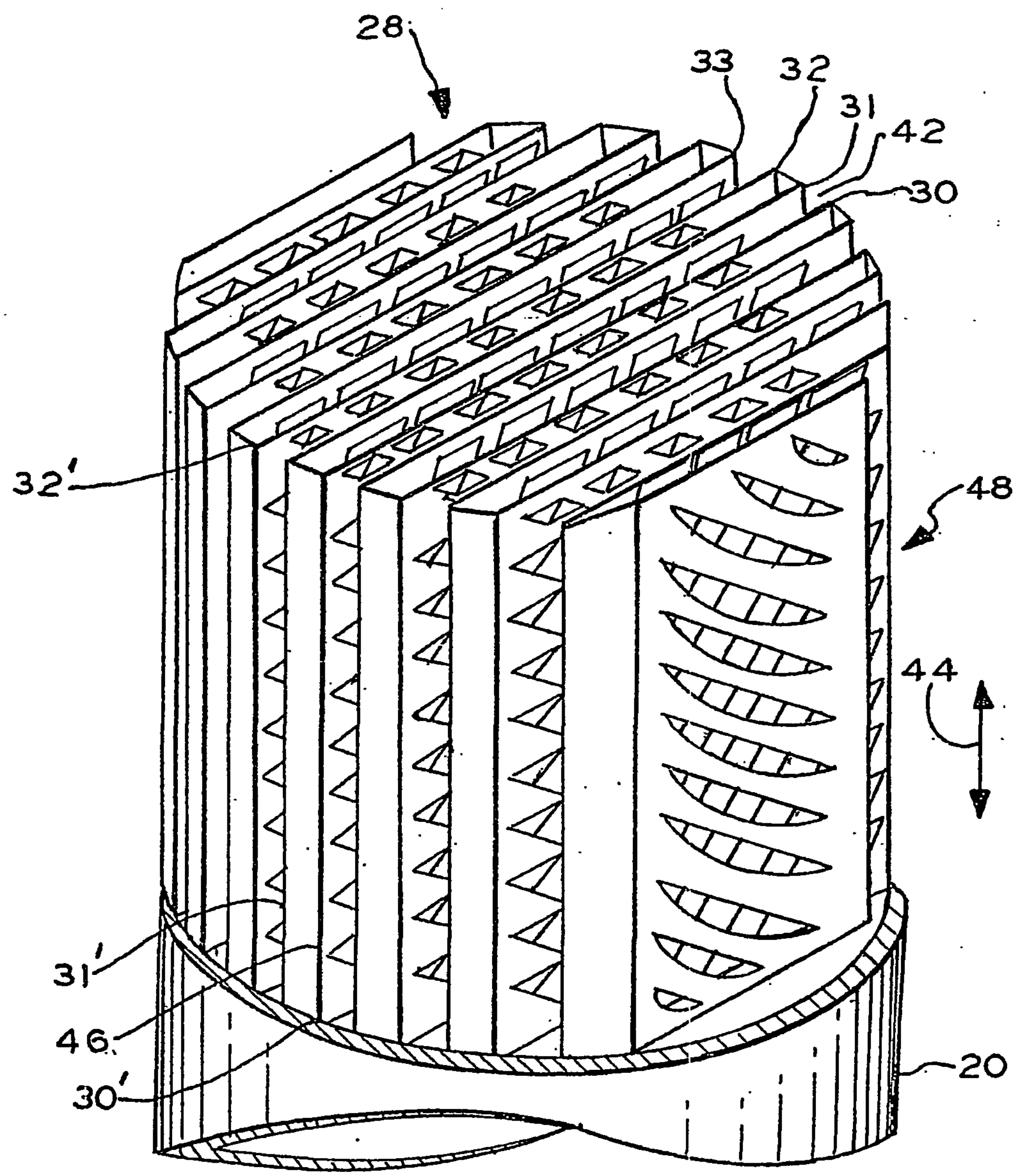
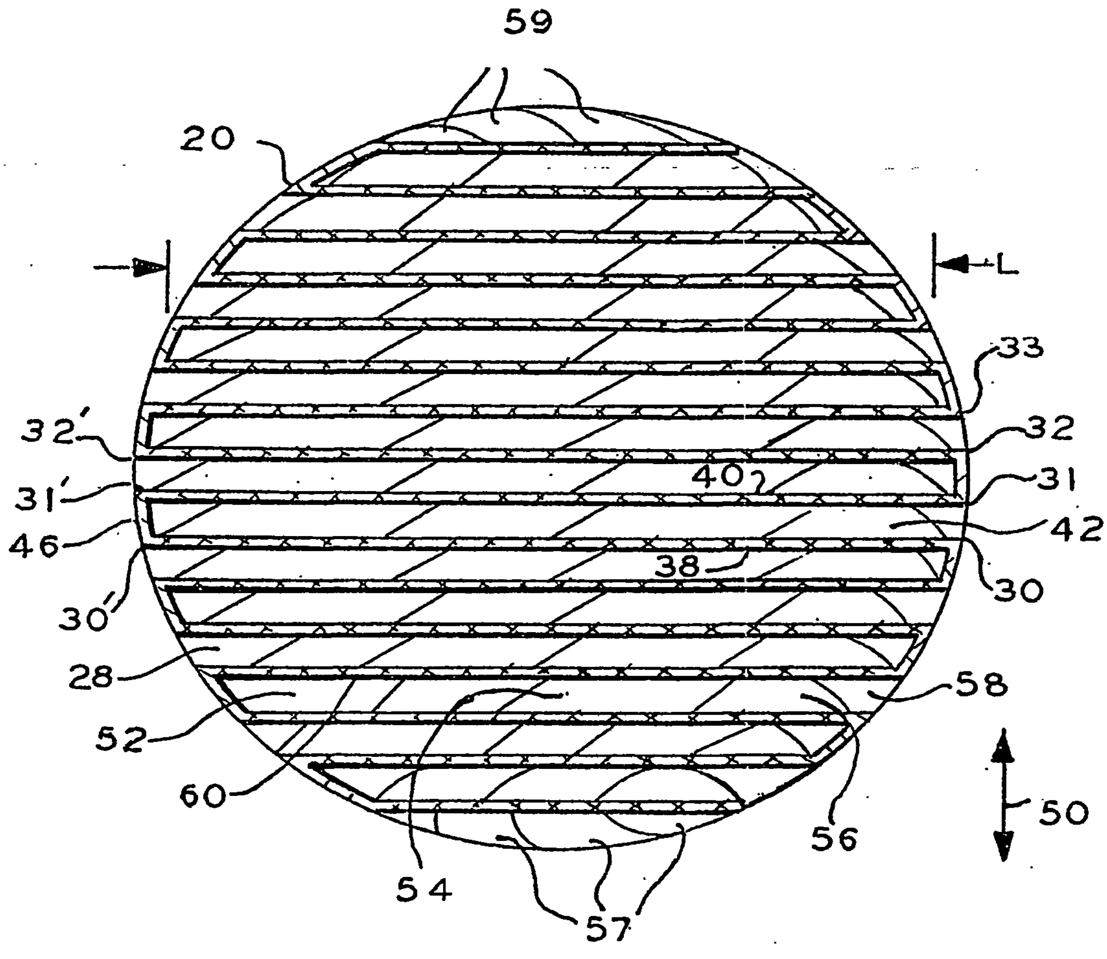


FIG. 4



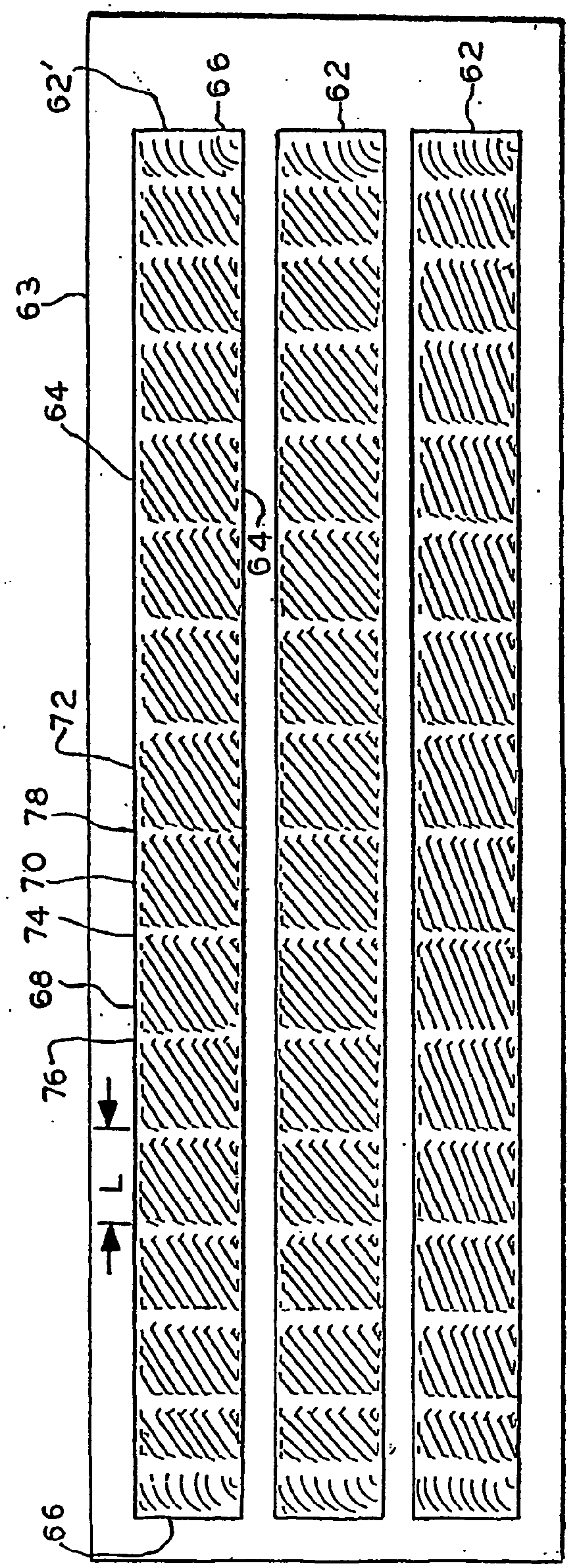


FIG. 5

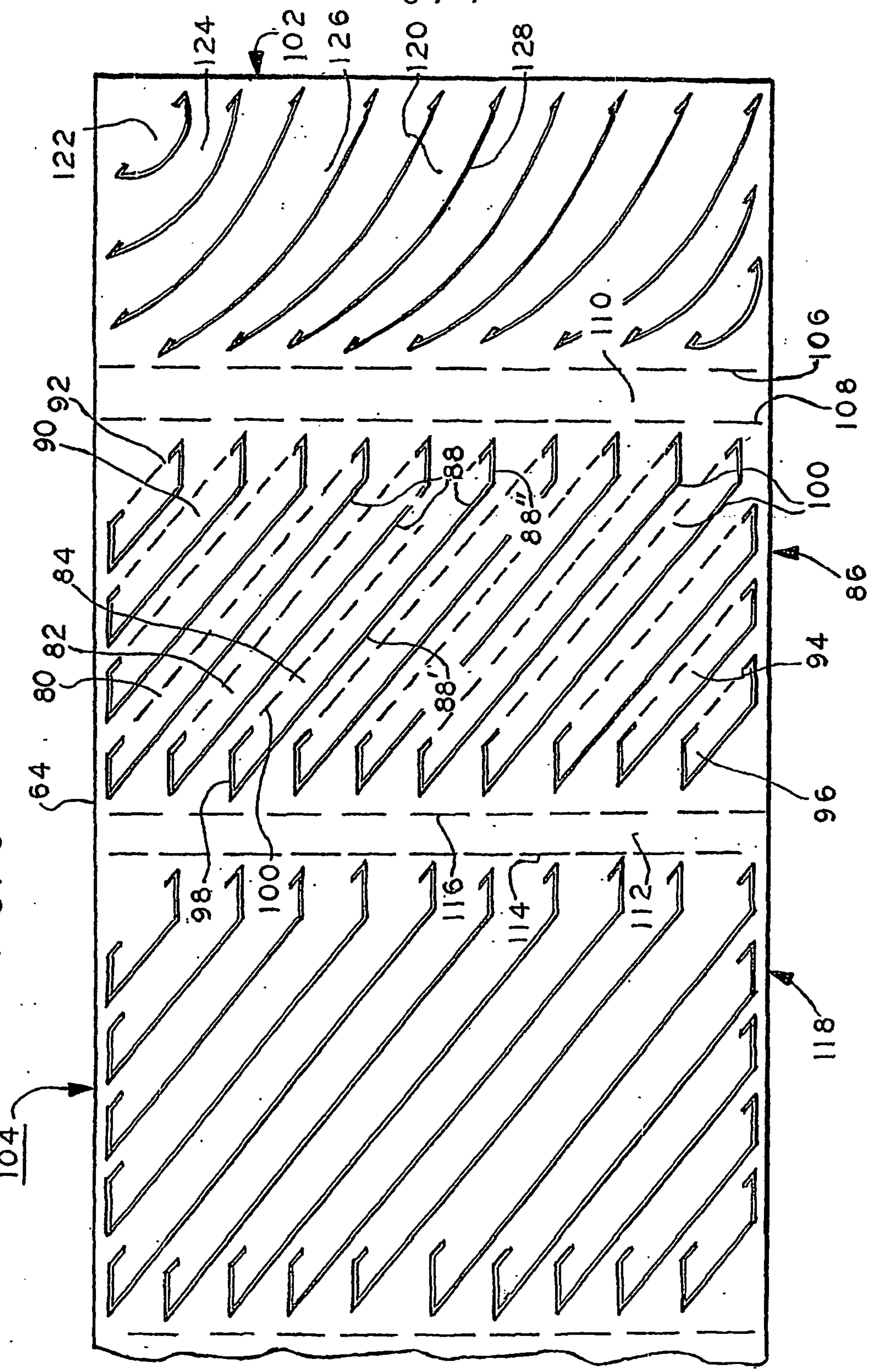

122
124
102
126
120
128
110
106
108
92
90
88
88'
88''
100
86
84
82
80
94
96
64
98
116
114
112
118
104

FIG. 6

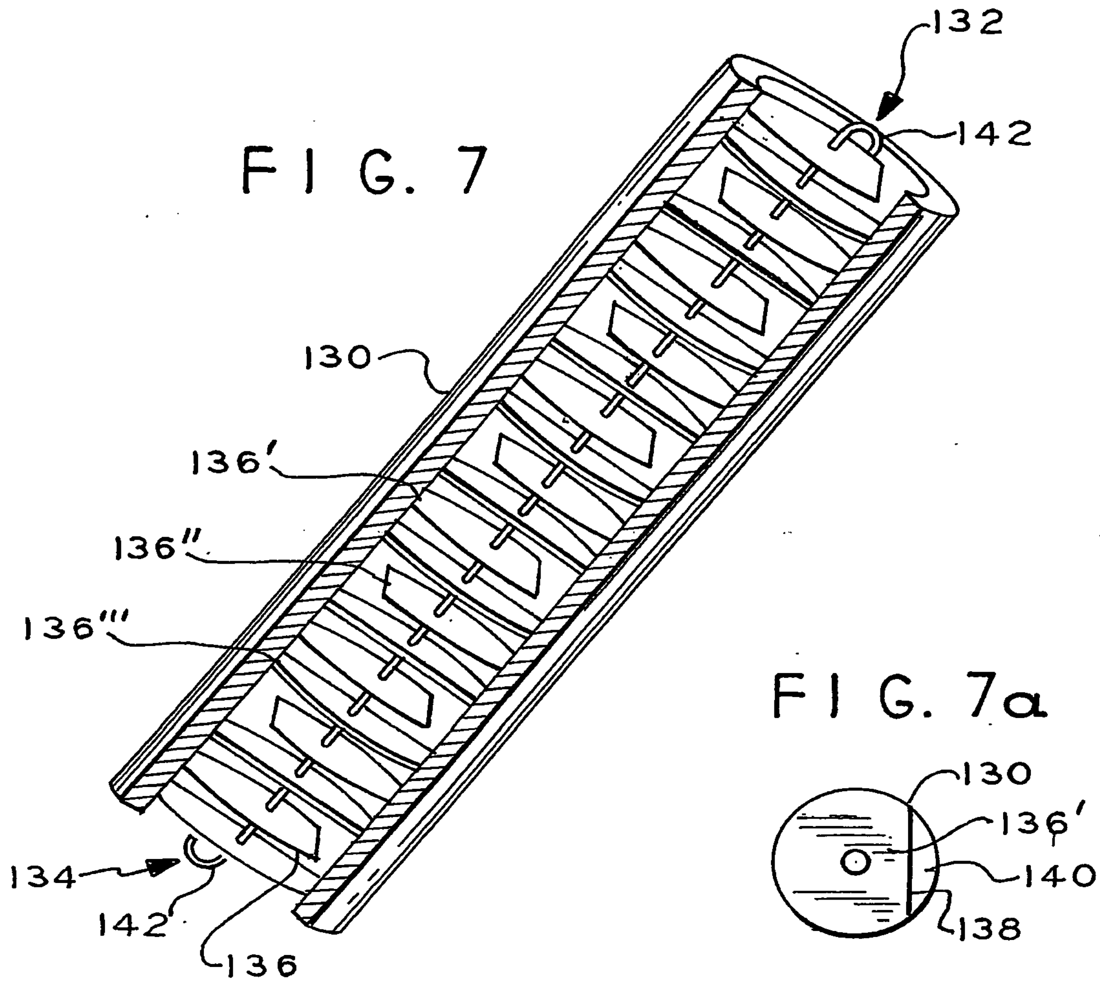


FIG. 7

FIG. 7a

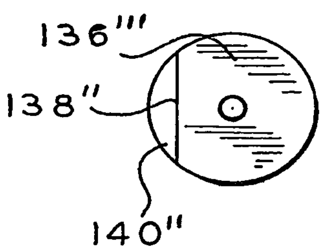


FIG. 7c

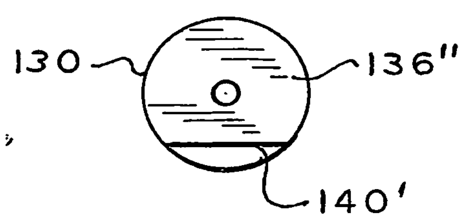


FIG. 7b